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STAGES IN Al PARTICLE COMBUSTION IN AIR

Edward L. Dreizin

AeroChem Research Laboratories, Inc., P.O. Box 12, Princeton, NJ 08542

BACKGROUND

Combustion of Al is of great practical interest and has been widely studied, e.g., [1]. Brightness oscillations and micro-explosions were observed in early Al particle combustion studies [1], however these processes are still not understood. Experiments on metal particle combustion conducted recently using a micro-arc device called GEnerator of Monodisperse MEtal Droplets (GEMMED) [2, 3] demonstrated that similar processes (brightness and temperature jumps, micro-explosions) are common in the combustion of a representative group of metals. It was suggested that such processes are caused by oxygen penetration into the burning metal particle and its subsequent reaction with metal, a process which is initiated when the metal-oxygen system reaches its typical temperature of non-variant transformation determined from the binary phase diagram.

Initial experiments on Al combustion using GEMMED [4] showed that voids are formed in burning Al particles and brightness fluctuations are observed during particle quenching in inert gas. It was suggested that, similarly to other metals, reactions occurring on the burning particle surface and (or) in its interior should be considered in order to understand the complete process of Al combustion.

The objective of this research is to determine experimentally the temperature histories of freely falling burning Al particles and consider them in conjunction with the Al-O phase diagram in order to determine possible relevant Al-O heterogeneous reactions.

EXPERIMENTAL

Al particle combustion experiments were conducted using GEMMED described elsewhere [5]. Uniform Al droplets of 120, 160, and 190 μm were produced and burned in air at atmospheric pressure.

Particle Temperature: the color temperature of the droplets was monitored using a three-wavelength pyrometer. The pyrometer included an iris, a fiber optics trifurcated bundle, three interference filters, and three HC120-01 Hamamatsu photo-sensor modules. The wavelengths of the interference filters used (520, 580, and 458 nm) were chosen so that no bands observed in the Al-O spectrum [6] could contribute to the measured signals. The pyrometer was calibrated using a tungsten strip lamp, providing a maximum black body temperature of 2650 K. The calibration showed that two intensity ratios (r_1=I(580 nm)/I(520 nm) and r_2=I(580 nm)/I(458 nm)) could be used for temperature measurements with an acceptable error (less than $\pm$80 K) in the temperature range 1900 - 2650 K.

The high sensitivity of the pyrometer allowed positioning far from the burning particle so that the entire particle trajectory was in the pyrometer field of view. Three brightness signals were recorded simultaneously for each particle combustion event and used to compute two color temperature histories. The temperatures above 2650 K were computed using an extrapolation of the calibration curve. The two color temperatures inferred for the Al combustion experiments agreed to $\pm$50 K in the range of 1900-3300 K, although the difference increased to +150 K (r_2 became higher) at higher temperatures.

Flame structure: Al is known to burn in the vapor-phase, and therefore, radiation from the burning gas surrounding the particle was expected to contribute significantly to the measured radiation. Spatial resolution of the measured optical signals from the particle and the flame zone was achieved using a screen positioned in front of the burning particle trajectory with a series of equally spaced, 0.2 mm wide slots perpendicular to the particle velocity vector. Radiation of burning particles moving behind the slots was registered as a series of pulses. The period of the pulses was used to determine the particle velocity, v, while the duration t_p of each pulse corresponded to the time needed to cross a slot. The size of the luminous domain was estimated as $v \cdot t_p - w$, where w is the slot width. Temperature profiles inferred from single pulses were assumed to correlate with real temperature profiles in the burning particle-flame complex.

Flame shape was visualized by a video-recording of the combustion events using a free running camera at fast shutter speed (0.5 ms exposure time). Also, particles were quenched on glass slides at different combustion times and the shape of smoke traces surrounding the quenched particles were examined.

Internal Particle Composition: particles were quenched at different combustion times by allowing them to pass into an argon atmosphere separated from the air by a soap-bubble film [3] or by impinging burning particles onto a flat plate of Cerrobend Alloy (melting point 90 °C). Cross-sections of particles quenched at different times were examined using SEM; the particle internal composition was studied using energy-dispersing spectroscopy (EDS).

RESULTS

The experimental temperature histories of the burning Al particles of different diameters were similar, the major difference being in the total combustion time. The combustion times were 50, 80, and 100 ms for particles of 120, 160, and 190 μm diameter, respectively. An example of one of the measured intensity signals (with no slots used) and one of the inferred temperatures is shown in Fig.1. Three distinct stages can be seen on all such curves.

The initial stage (0-25 ms for the particles of 190 μm diameter, as one can see from Fig.1) consists of an increased and then stabilized radiation intensity and quite similar temperature variation. The temperature stabilizes at approximately 2900 °C which is close to the Al_2O_3 boiling point (2980 °C) and exceeds the boiling point of Al (2520 °C). This implies an envelope flame contribution which is confirmed with spatially resolved measurements. An example of a spatially resolved intensity pulse and corresponding temperature curve registered for a particle crossing the slot during this initial combustion stage is shown in Fig. 2a. This demonstrates a spherically-symmetric combustion, with a central region temperature close to the Al_2O_3 boiling point and higher temperatures close to the luminous zone edges. It can be understood assuming that the vapor phase Al-O_2 reaction produces gaseous products (sub-oxides) which tend to form condensed stoichiometric Al_2O_3 in a cooler area close to the particle surface where temperature is limited by the Al boiling point. Fig. 2a indicates that the high-temperature gas-phase reaction zone is approximately 150 μm away from the particle surface. Similar conclusions related to spherically symmetric flame and combustion zone diameter, can be drawn from the shape of the smoke cloud surrounding quenched particle on a glass slide.

The second stage (see Fig. 1, 25-45 ms) can be characterized by a remarkably increased intensity of the measured radiation, and initiation of its strong oscillations. Smoke traces around the quenched particles demonstrate a transition to a non-symmetric combustion, increase of the smoke cloud size, and approaching the reaction zone to the particle surface. This is again consistent with spatially resolved optical measurements. An intensity pulse and corresponding temperature profile for the particle crossing a slot during this stage is shown in Fig. 2b. The intensity pulse shows a bi-modal structure. The temperature profile shows the presence of at least two reaction zones. The radiation from the particle surface expected at the beginning of the signal is shielded by the oscillating radiation of a smoke cloud, existing close to the surface. A slight decrease in the temperature measured without slots is observed at this time.

The third, final, combustion stage (45-100 μs in Fig. 1) is observed during which the intensity of the measured radiation signal decreases and then remains nearly constant on the background of repeatable oscillations. The temperature is also observed to decrease during this stage, but this decrease is observed after the radiation decrease, while radiation intensity does not change considerably. The smoke clouds around the quenched particles collected under third stage conditions continue to grow and show non-symmetric structure. Particle velocity vector changes noticeably during this third stage and, as a rule the combustion is terminated by a micro-explosion (documented on a video-tape).

EDS analysis of the internal composition of Al particles quenched during the combustion shows presence of oxygen. Its concentration increases with increasing the combustion time.

DISCUSSION

The temperature measurements indicate that Al metal boils during the combustion. The processes occurring during the first observed stage apparently are similar to the classical description of vapor-phase metal combustion. An important detail is that oxygen is already found in the Al particles quenched during this stage. A realistic assumption is that suboxides (AlO and AlO_2) formed in the reaction zone diffuse to (as well as from) the particle, and a fraction of these gaseous suboxides dissolve in the liquid Al.

The binary phase diagram of Al-O system [7] suggests important changes in the Al-O system when the concentration of dissolved oxygen reaches approximately 14% (atomic). The boiling point of Al slightly decreases (ca. 180 °C) and a liquid Al_2O_3 phase forms. A new composition for the vapor phase (containing vapor produced by Al_2O_3) appears at temperatures above the boiling point (which exists outside the particle, close to the gas-phase reaction zone). It is suggested that the transition from the first to the second combustion stage occurs when the concentration of oxygen dissolved in the Al particle reaches the 14% limit. The new vapor phase composition has a lower than pure Al pressure at the same temperatures, which is consistent with the observed approaching the reaction zone towards the particle surface. The particle itself remains close to the Al boiling point, however it consists of two liquids: almost pure Al and Al_2O_3. Such a composition can explain a non-symmetric combustion mode and the existence of local micro-flames.

The processes occurring during the non-symmetric combustion (second and third stages) can be explored using the shape of the smoke traces of the quenched particles. The smoke patterns observed during the second and third stages are remarkably similar to the electric field potential lines for a particle system having a spatial charge or a dipole moment. This implies that electric field effects play a significant role in the formation of the flame structure. In fact, charging (or polarizing) of the burning metal droplets could have resulted in their explosions. Such processes need to be considered in greater detail to understand nature of the third stage of Al combustion and micro-explosion phenomenon.

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REFERENCES

1. Naval Weapons Center. Metal Particle Combustion Progress Report, 1 July 1965-May 1967, by Metal Combustion Study Group. (J.L Prentice, Ed.). *NWC TP 4435*, China Lake CA, (1968)
2. Dreizin, E.L., Suslov, A.V. and Trunov, M.A. *Combust. Sci. Tech.* 87:45-58 (1992)
3. Dreizin, E.L., Suslov, A.V. and Trunov, M.A. *Combust. Sci. Tech.* 90:79-99 (1993)
4. Dreizin, E.L. and Trunov, M.A., submitted in *Combustion and Flame*, (1994).
5. Suslov, A.V., Dreizin, E.L. and Trunov, M.A., *Powder Tech.*, 74, 23 (1993)
6. Pearse, R.W.B., and Gaydon, A.G., *The Identification of Molecular Spectra*, Halsted Press, a Division of John Wiley & Sons, Inc., NY (1976)
7. Levinskiy, Y.V., *P-T-X Binary Phase Diagrams of Metal Systems*, Moscow, Metallurgia, (1990) (In Russian)